Dynamics of a metastable state nonlinearly coupled to a heat bath driven by an external noise

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Based on a system-reservoir model, where the system is nonlinearly coupled to a heat bath and the heat bath is modulated by an external stationary Gaussian noise, we derive the generalized Langevin equation with space dependent friction and multiplicative noise and construct the corresponding Fokker-Planck equation, valid for short correlation time, with space dependent diffusion coefficient to study the escape rate from a metastable state in the moderate to large damping regime. By considering the dynamics in a model cubic potential we analyze the result numerically which are in good agreement with the theoretical prediction. It has been shown numerically that the enhancement of rate is possible by properly tuning the correlation time of the external noise.

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I. INTRODUCTION

Barrier crossing phenomena are ubiquitous and are a central issue in many areas of natural science [1, 2]. Since the reaction coordinate describing the transition across the barrier typically interact with a large number of microscopic degrees of freedom, a useful theory to start with is the construction of a Hamiltonian which incorporates the effects of a heat bath environment. The classical treatment of thermally activated barrier crossing description is naturally accounted for by the generalized Langevin equation (may be nonlinear) and by the corresponding Fokker-Planck equation. Following Kramers [3, 4] transition rate can then be calculated from a nonequilibrium steady state solution of the Fokker-Planck equation describing a constant flux across the potential barrier. Over several decades the Kramers' theory and many of its variants has served as standard paradigms in various problems of physical and chemical kinetics to understand the rate in multidimensional systems in the overdamped and underdamped limits [5, 6], effects of anharmonicities [7], rate enhancement by parametric fluctuations [8], the role of non-Gaussian white noise [7, 9], role of a relaxing bath [10], quantum and semiclassical corrections to classical rate processes [11, 12, 13] and related similar aspects.

The common feature of overwhelming majority of the aforesaid treatments is that the system is thermodynamically closed, which means that the noise of the medium is of internal origin so that the dissipation and fluctuations gets balanced through the fluctuation-dissipation relation. However, in a number of situations the system is thermodynamically open, *i.e.*, when the system is driven by an external noise which is independent of system's characteristic damping [14]. The main feature of the dynamics in this case is the absence of any fluctuation-dissipation relation. While in the former case a zero current steady state situation is characterized by an equilibrium Boltzmann distribution, the corresponding situation in the later case is defined only by a steady state condition, if attainable.

A common approach to study the nonlinear, nonequilibrium systems involves a description in terms of nonlinear stochastic differential equation [15]. From a microscopic point of view, the system-reservoir Hamiltonian description suggests that the coupling of the system and the reservoir coordinates determines both the noise and the dissipative terms in the Langevin equation describing the dynamics of the system. If the system-bath interaction is linear in the bath coordinates but arbitrary in the system coordinate, the corresponding generalized Langevin equation incorporates a multiplicative noise and consequently a nonlinear dissipative term arises due to the nonlinear system-bath interaction [16]. A canonical distribution for initial conditions of the bath variables yields a zero average for the fluctuations and the fluctuation-dissipation relation is again maintained [16]. However, when the reservoir is modulated by an external noise, it is likely that it induces fluctuations in the polarization of the reservoir [17]. Since the fluctuations of the reservoir are crucially dependent on the response function, one can envisage a connection between the dissipation of the system and the response function of the reservoir due to the external noise from a microscopic stand point [17]. A direct driving of the system usually breaks the fluctuation-dissipation relation and can generate a biased directed motion that are seen in ratchet and molecular motors [18]. On the other hand the bath modulation by an external noise agency maintains a thermodynamic consistency relation, an analogue of the fluctuation-dissipation relation of the closed system, as a result of which the well known Kramers' turnover feature

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can be restored [17].

While the nonequilibrium, nonthermal systems have also been investigated phenomenologically by a number of works in several contexts [19], these treatment are mainly concerned with direct driving of the system by an external noise or a time dependent field. In the present paper we consider a system-reservoir model where the reservoir is modulated by an external noise and the system is nonlinearly coupled to a heat bath thereby resulting a nonlinear, multiplicative generalized Langevin equation. Our object here is to explore the role of reservoir response on the system's dynamics and to calculate the generalized escape rate from a metastable state for a nonequilibrium open system in the presence of multiplicative noise.

A number of different situations depicting the modulation of the heat bath may be physically relevant. As for example, one may experimentally study the reversible isomerization of *cis*-butene to *trans*-butene, *cis*-butene $\rightleftharpoons$ *trans*-butene. In terms of the reaction rate theory both isomers represent the two stable local minima of the potential energy landscape and are separated by the activation energy which one stable configuration (*cis* or *trans*) needs to overcome to be converted into its isomeric form. To observe the effect of external stochastic modulation one can carry out the experiment in a photochemically active solvent (the heat bath) where the solvent is under the influence of external monochromatic light with fluctuating intensity of a wavelength which is absorbed solely by the solvent molecules. As a result of it the modulated solvent heats up due to the conversion of light energy into heat energy by radiationless relaxation process and a effective temperature like quantity develops due to the constant input of energy. Since the fluctuations in the light intensity results in the polarization of the solvent molecules, the effective reaction field around the reactants gets modified. Provided the required stationarity of this nonequilibrium open system is maintained the dynamics of the barrier crossing becomes amenable to the present theoretical analysis that follows.

Though the dynamics of a Brownian particle in a uniform solvent is well-known, it is not very clear when the response of the solvent becomes time dependent, as in a liquid crystal when projected onto an anisotropic stochastic equation of motion or in the diffusion and reaction in supercritical liquids and growth in living polymerization [20, 21]. Also space-dependent friction is realized from the presence of a stochastic potential in the Langevin equation [22]. An exact Fokker-Planck equation for time and space dependent friction was derived by Pollak *et al* [23]. Along with these formal developments, the theories of multiplicative noise have found in wide applications in several areas, *e.g.*, activated rate processes, stochastic resonance, laser and optics, noise induced transport etc [24]. In passing we mention that the escape rate for a space dependent friction is just not a theoretical issue but has been a subject of experimental investigation over the last two decades [25].

The organization of the paper is as follows: In section II, starting from a Hamiltonian description of a system nonlinearly coupled to a harmonic reservoir which is modulated externally by a Gaussian noise, we have derived the generalized Langevin equation with an effective Gaussian noise $\xi(t)$. The statistical properties of $\xi(t)$ has also been explored. In section III we have constructed the corresponding Fokker-Planck equation, valid for small correlation time, and have derived the generalized Kramers' rate for moderate to large friction. In section IV, a specific example has been carried out. Both the numerical and analytical results have been analyzed in section V. The paper has been concluded in section VI.

II. THE MODEL: HEAT BATH MODULATED BY EXTERNAL NOISE

We consider a classical particle of mass M nonlinearly coupled to a heat bath consisting of N harmonic oscillators driven by an external noise. The total Hamiltonian of such a composite system can be written as [26, 27]

$$H = \frac{p^2}{2M} + V(x) + \frac{1}{2}\sum_{j=1}^{N}\left\{\frac{p_j^2}{m_j} + m_j\omega_j^2\left(q_j - c_j g(x)\right)^2\right\} + H_{int} \tag{2.1}$$

where x and p are the coordinate and the momentum of the system particle, respectively, and $V(x)$ is the potential energy of the system. (q_j, p_j) are the variables for the jth bath oscillator having frequency ω_j and mass m_j. c_j is the coupling constant for the system-bath interaction and $g(x)$ is some analytic function of system coordinate. H_{int} is the interaction term between the heat bath and the external noise $\epsilon(t)$, with the following form:

$$H_{int} = \sum_{j=1}^{N}\kappa_j q_j \epsilon(t). \tag{2.2}$$

The type of interaction we have considered between the heat bath and the external noise, H_{int} is commonly known as the *dipole interaction* [28]. In equation (2.2), κ_j denotes the strength of the interaction. We consider $\epsilon(t)$ to be a stationary, Gaussian noise process with zero mean and arbitrary correlation function

$$\langle\epsilon(t)\rangle_e = 0, \qquad \langle\epsilon(t)\epsilon(t')\rangle_e = 2D\Psi(t-t') \tag{2.3}$$

where D is the external noise strength, $\Psi(t-t')$ is the external noise kernel and $\langle ...\rangle_e$ implies averaging over the external noise processes.

Eliminating the bath degrees of freedom in the usual way [16] (and setting M and $m_j = 1$) we obtain the generalized Langevin equation

$$\begin{aligned}\dot{x} &= v \\ \dot{v} &= -\frac{dV(x)}{dx} - \frac{dg(x)}{dx}\int_0^t dt'\gamma(t-t')\frac{dg(x(t'))}{dx(t')}v(t') \\ &\quad + \frac{dg(x)}{dx}\{f(t)+\pi(t)\} \end{aligned} \tag{2.4}$$

where

$$\gamma(t) = \sum_{j=1}^{N} c_j^2\omega_j^2 \cos\omega_j t \tag{2.5}$$

and $f(t)$ is the thermal fluctuation generated due to the system-reservoir interaction and is given by

$$\begin{aligned} f(t) &= \sum_{j=1}^{N}\Big[c_j\omega_j^2\{q_j(0) - c_j g(x(0))\}\cos\omega_j t \\ &\quad + \frac{v_j(0)}{\omega_j}\sin\omega_j t\Big]. \end{aligned} \tag{2.6}$$

In equation (2.4), $\pi(t)$ is the fluctuating force generated due to the external stochastic driving $\epsilon(t)$ and is given by

$$\pi(t) = -\int_0^t dt'\varphi(t-t')\epsilon(t') \tag{2.7}$$

where

$$\varphi(t) = \sum_{j=1}^{N} c_j\omega_j\kappa_j \sin\omega_j t. \tag{2.8}$$

The form of equation (2.4) indicates that the system is driven by two forcing terms $f(t)$ and $\pi(t)$, both are multiplicative by a function of system variable $dg(x)/dx$. Thus we have obtained a generalized Langevin equation with multiplicative noise. To define the statistical properties of $f(t)$, we assume that the initial distribution is one in which the bath is equilibrated at $t = 0$ in the presence of the system but in the absence of the external noise agency $\epsilon(t)$ such that

$$\langle f(t)\rangle = 0 \text{ and } \langle f(t)f(t')\rangle = k_B T\gamma(t-t').$$

Now at $t = 0_+$, the external noise agency is switched on and the bath is modulated by $\epsilon(t)$. The system dynamics is governed by equation (2.4), where apart from the internal noise $f(t)$ another fluctuating force $\pi(t)$ appears that depends on the external noise $\epsilon(t)$. So we define an effective noise $\xi(t)[= f(t)+\pi(t)]$ whose correlation is given by [17]

$$\begin{aligned}\langle\langle\xi(t)\xi(t')\rangle\rangle &= k_B T\gamma(t-t') + 2D\int_0^t dt''\int_0^{t'} dt''' \\ &\quad \times\varphi(t-t'')\varphi(t'-t''')\Psi(t''-t''')\end{aligned} \tag{2.9}$$

along with $\langle\langle\xi(t)\rangle\rangle = 0$, where $\langle\langle ...\rangle\rangle$ means we have taken two averages independently. It should be noted that the above equation (2.9) is not a fluctuation-dissipation relation due to the appearance of the external noise intensity. Rather it serves as a thermodynamic consistency condition. The statistical properties of $\pi(t)$ are determined by the normal mode densities of the bath frequencies, the coupling of the system with the bath, the coupling of the bath with the external noise and on the statistical properties of the external noise itself. Equation (2.7) is reminiscent of the familiar linear relation between the polarization and external field, where $\pi(t)$ and $\epsilon(t)$ play the role of the former and later, respectively. $\varphi(t)$ then can be interpreted as a response function of the reservoir due to the external noise $\epsilon(t)$. The structure of $\pi(t)$ suggests that this forcing function, although obtained from an external agency, is different from a direct driving force acting on the system.

To obtain a finite result in the continuum limit, the coupling function $c_i = c(\omega)$ and $\kappa_i = \kappa(\omega)$ are chosen [17] as $c(\omega) = c_0/\omega\sqrt{\tau_c}$ and $\kappa(\omega) = \kappa_0\omega\sqrt{\tau_c}$. Consequently $\gamma(t)$ and $\varphi(t)$ reduces to the following forms:

$$\gamma(t) = \frac{c_0^2}{\tau_c}\int d\omega\mathscr{D}(\omega)\cos\omega t \tag{2.10}$$

and

$$\varphi(t) = c_0\kappa_0\int d\omega\mathscr{D}(\omega)\omega\sin\omega t \tag{2.11}$$

where c_0 and κ_0 are constants and $1/\tau_c$ is the cutoff frequency of the oscillator. τ_c may be characterized as the correlation time of the bath [16]. For $\tau_c \to 0$ we obtain a delta-correlated noise process. $\mathscr{D}(\omega)$ is the density of modes of the heat bath which is assumed to be Lorentzian:

$$\mathscr{D}(\omega) = \frac{2}{\pi}\frac{1}{\tau_c(\omega^2+\tau_c^{-2})}. \tag{2.12}$$

This assumption resembles broadly the behavior of the hydrodynamical modes in a macroscopic system [20]. With these forms of $\mathscr{D}(\omega)$, $c(\omega)$ and $\kappa(\omega)$ we have the expression for $\varphi(t)$ and $\gamma(t)$ as

$$\varphi(t) = \frac{c_0\kappa_0}{\tau_c}\exp(-t/\tau_c) \tag{2.13a}$$

$$\gamma(t) = \frac{c_0^2}{\tau_c}\exp(-t/\tau_c). \tag{2.13b}$$

From equations (2.10) and (2.11) we obtain

$$\frac{d\gamma(t)}{dt} = -\frac{c_0}{\kappa_0}\frac{1}{\tau_c}\varphi(t), \tag{2.14}$$

Equation (2.14), an important content of the present model, is independent of $\mathscr{D}(\omega)$. This expresses how the dissipative kernel $\gamma(t)$ depends on the response function $\varphi(t)$ of the medium due to the external noise $\epsilon(t)$.

If we assume that $\epsilon(t)$ is a δ-correlated noise, *i.e.*, $\langle\epsilon(t)\epsilon(t')\rangle_e = 2D\delta(t-t')$ then the correlation function of $\pi(t)$ will be

$$\langle\pi(t)\pi(t')\rangle = \frac{Dc_0^2\kappa_0^2}{\tau_c}\exp(-|t-t'|/\tau_c) \tag{2.15}$$

where we have neglected the transient terms $(t, t' > \tau_c)$. This equation shows how the heat bath dresses the external noise. Though the external noise is a δ-correlated noise, the system encounters it as an Ornstein-Uhlenbeck noise with same correlation time of the heat bath but with an intensity depending on the coupling κ_0 and the external noise strength D. On the other hand, if the external noise is an Ornstein-Uhlenbeck process with $\langle\epsilon(t)\epsilon(t')\rangle_e = (D/\tau')\exp(-|t-t'|/\tau')$, the correlation function of $\pi(t)$ is found to be

$$\begin{aligned}\langle\pi(t)\pi(t')\rangle &= \frac{Dc_0^2\kappa_0^2}{(\tau'/\tau_c)^2-1}\frac{\tau'}{\tau_c}\left\{\frac{1}{\tau_c}\exp\left(-\frac{|t-t'|}{\tau'}\right)\right.\\ &\left.-\frac{1}{\tau'}\exp\left(-\frac{|t-t'|}{\tau_c}\right)\right\}\end{aligned} \tag{2.16}$$

where we have neglected the transient terms. If the external noise-correlation time be much larger than the internal noise-correlation time, *i.e.*, $\tau' \gg \tau_c$, which is more realistic, then the dressed noise is dominated by the external noise and we have from (2.16)

$$\langle\pi(t)\pi(t')\rangle = \frac{Dc_0^2\kappa_0^2}{\tau'}\exp\left(-\frac{|t-t'|}{\tau'}\right). \tag{2.17}$$

On the other hand, when the external noise correlation time is smaller than the internal one, we recover (2.15).

III. GENERALIZED FOKKER-PLANCK DESCRIPTION AND KRAMERS' ESCAPE RATE

To start with we consider the internal dissipation is Markovian (*i.e.* $\tau_c \to 0$ and the internal noise is Gaussian δ-correlated)

$$\gamma(t) = 2\beta\delta(t-t') \text{ where } \beta = c_0^2. \tag{3.1}$$

Consequently the generalized Langevin equation (2.4) reduces to

$$\begin{aligned}\dot{x} &= v\\ \dot{v} &= -\frac{dV(x)}{dx} - \beta(g'(x))^2 v + g'(x)\{f(t)+\pi(t)\}\\ &= -\frac{dV(x)}{dx} - \Gamma(x)v + g'(x)\{f(t)+\pi(t)\}\end{aligned} \tag{3.2}$$

where

$$\Gamma(x) = \beta(g'(x))^2. \tag{3.3}$$

Using van Kampen's cumulant expansion method [29], the Fokker-Planck equation, valid for small correlation time, corresponding to the above generalized Langevin equation is obtained as (see Appendix A)

$$\begin{aligned}\frac{\partial P(x,v,t)}{\partial t} &= -v\frac{\partial P}{\partial x}\\ &+[\Gamma(x)v + V'(x) - 2g'(x)g''(x)J_e]\frac{\partial P}{\partial v}\\ &+A\frac{\partial^2 P}{\partial v^2} + B\frac{\partial^2 P}{\partial v\partial x} + \Gamma(x)P\end{aligned} \tag{3.4}$$

where

$$A = (g'(x))^2 I_e - \Gamma(x)(g'(x))^2 J_e \text{ and } B = (g'(x))^2 J_e, \tag{3.5}$$

and I_e and J_e are defined as

$$I_e = \int_0^\infty \langle\xi(t)\xi(t-\tau)\rangle d\tau, \tag{3.6a}$$

$$J_e = \int_0^\infty \tau\langle\xi(t)\xi(t-\tau)\rangle d\tau. \tag{3.6b}$$

In equations (3.6a-3.6b), $\xi(t)$ is the effective noise term $[\xi(t) = f(t)+\pi(t)]$ as defined earlier. In deriving (3.4) we have assumed that $f(t)$ and $\epsilon(t)$ are uncorrelated as they have different origin. *Equation (3.4) is the first key result of this paper.* It should be noted that when the noise is purely internal and the system-reservoir coupling is linear, equation (3.4) reduces to the generalized Kramers equation [4] (valid for small correlation time).

In Kramers' original treatment the dynamics of the Brownian particle was governed by Markovian random processes. Since the work of Kramers a number of authors [30, 31, 32, 33] have extended Kramers' analysis for the non-Markovian case to derive the expression for generalized escape rate. In order to allow ourselves a comparison with Fokker-Planck equation of other forms [31, 32, 34], we note that the diffusion coefficient in equation (3.4) is coordinate dependent. It is customary to get rid of this coordinate dependence by approximating the coefficients at the barrier top or potential well where we need the steady state solution of equation (3.4). One may also use mean field solution of equation (3.4) obtained by

neglecting the fluctuation terms and putting appropriate stationary condition in the diffusion coefficient. The drift term in equation (3.4) refers to the presence of a dressed potential of the form

$$R(x) = V(x) - (g'(x))^2 J_e. \tag{3.7}$$

The modification of the potential is essentially due to nonlinear coupling of the system to the nonequilibrium modes. J_e is a non-Markovian small contribution and therefore the second term of the above equation may be neglected for small correlation time. For the rest of the treatment we use $R(x) \simeq V(x)$. For a harmonic oscillator with frequency ω_0, $V(x) = \omega_0^2 x^2/2$; the linearized version of Fokker-Planck equation is represented as

$$\frac{\partial P}{\partial t} = -v\frac{\partial P}{\partial x} + \Gamma P + [\Gamma v + \omega_0^2 x]\frac{\partial P}{\partial v} + A_0\frac{\partial^2 P}{\partial v^2} + B_0\frac{\partial^2 P}{\partial v \partial x} \tag{3.8}$$

where

$$A_0 = (g'(0))^2 I_e - \Gamma(0)(g'(0))^2 J_e \text{ and } B_0 = (g'(0))^2 J_e \tag{3.9}$$

are calculated at the bottom of the potential ($x = 0$). From equation (3.9) we have

$$A_0 = (g'(0))^2 I_e - \Gamma(0) B_0. \tag{3.10}$$

The general steady state solution of equation (3.8) becomes

$$P_{st}(x, v) = \frac{1}{Z}\exp\left[-\frac{v^2}{2D_0} - \frac{\omega_0^2 x^2}{2(D_0 + B_0)}\right] \tag{3.11}$$

where

$$D_0 = \frac{A_0}{\Gamma(0)} \tag{3.12}$$

and Z is the normalization constant. The solution (3.11) can be verified by direct substitution in the steady state ($\partial P(x = 0, v)/\partial t = 0$) version of the Fokker-Planck equation (3.4), namely

$$-v\frac{\partial P_{st}}{\partial x} + \Gamma P_{st} + [\Gamma v + \omega_0^2 x]\frac{\partial P_{st}}{\partial v} + A_0\frac{\partial^2 P_{st}}{\partial v^2} + B_0\frac{\partial^2 P_{st}}{\partial v \partial x} = 0 \tag{3.13}$$

The distribution (3.11) is not an equilibrium distribution. In absence of the external noise $\epsilon(t)$ it reduces to the standard thermal Boltzmann distribution, $\exp[-(v^2 + V(x))/k_B T]$. Thus the steady state distribution for the nonequilibrium open system plays the role of an equilibrium distribution of the closed system which may however be recovered in the absence of the external noise.

We now turn to the problem of decay of a metastable state. In Kramers' approach [3], the particle coordinate x corresponds to the reaction coordinate, and its values at the minima of the potential well $V(x)$ separated by a potential barrier, denotes the reactant and product states.

Linearizing the motion around the barrier top at $x = x_b$, the steady state ($\partial P(x = x_b, v)/\partial t = 0$) Fokker-Planck equation corresponding to equation (3.4) reads

$$-v\frac{\partial P_{st}}{\partial y} - \omega_b^2 y\frac{\partial P_{st}}{\partial v} + \Gamma(x_b)\frac{\partial}{\partial v}(vP_{st}) + A_b\frac{\partial^2 P_{st}}{\partial v^2} + B_b\frac{\partial^2 P_{st}}{\partial v \partial y} = 0, \tag{3.14}$$

where

$$y = x - x_b,\ V(y) = E_b - \frac{1}{2}\omega_b^2 y^2,\ \omega_b^2 > 0 \tag{3.15}$$

and the suffix 'b' indicates that all the coefficients are to be calculated using the general definition of A and B (3.5) at the barrier top. It is interesting to note that for linear coupling, we can extend our analysis for arbitrary correlation time and in such a case, the barrier dynamics would have been governed by the Fokker-Planck equation of Adelman's form [34].

To derive a nonvanishing diffusion current across the barrier top Kramers [3] considered $P_b(x, v)$ to be the equilibrium Boltzmann distribution ($\exp[-(v^2 + V(x))/k_B T]$) multiplied by a propagator $F(x, v)$ and used it to solve the Fokker-Planck equation. In our model the equilibrium distribution should be replaced by the steady state distribution which depends on the local nature of the potential at the barrier top and the effective temperature like quantity for the nonequilibrium open system. However, in the absence of the external noise $\epsilon(t)$ the steady state distribution reduces to the equilibrium Boltzmann distribution. Following Kramers [3], we thus assume that the nonequilibrium steady state probability $P_b(x, v)$ generating a nonvanishing diffusion current across the barrier is given by

$$P_b(x, v) = \exp\left[-\left\{\frac{v^2}{2D_b} + \frac{V(x)}{D_b + B_b}\right\}\right] F(x, v), \tag{3.16}$$

where

$$D_b = \frac{A_b}{\Gamma(x_b)}, \tag{3.17}$$

with

$$\begin{aligned} V(x) &= E_0 + \frac{1}{2}\omega_0^2 x^2, \text{ near the bottom} \\ &= E_b + \frac{1}{2}\omega_b^2 (x - x_b)^2, \text{ near the top.} \end{aligned}$$

The expression (3.16) denoting the steady state distribution is motivated by the local analysis near the bottom and the top of the potential. For a stationary nonequilibrium system, on the other hand, the relative population of the two regions, in general, depends on the global properties of the potential leading to an additional factor in the rate expression. Because of the Kramers' type ansatz, which is valid for the local analysis, such a consideration is outside the scope of the present treatment.

Following Kramers' original reasoning [3] we then derive the barrier crossing rate K, for moderate to large friction regime (see Appendix B for detailed calculation)

$$K = \frac{\omega_0}{2\pi}\frac{D_b}{(D_0+B_0)^{1/2}}\left(\frac{\Lambda}{1+\Lambda D_b}\right)^{1/2}\exp\left(\frac{-E_b}{D_b+B_b}\right) \tag{3.18}$$

where E_b is the barrier height of the potential and the parameter Λ is given by

$$\Lambda = \frac{\lambda}{A_b + aB_b}$$

with

$$\begin{aligned} -\lambda &= \Gamma(x_b) + a\left(1+\frac{B_b}{D_b}\right), \\ a &= \frac{D_b}{2(D_b+B_b)}\left\{-\Gamma(x_b) - \sqrt{\Gamma^2(x_b)+4\omega_b^2}\right\}. \end{aligned}$$

The strength of the external noise and the damping function are buried in the parameters D_0, B_0, D_b, B_b and Λ. Equation (3.18) is *the second key result of this paper*. Here we note that $(D_b + B_b)/k_B$ in the exponential factor of (3.18) defines a new effective temperature characteristic of the steady state of the nonequilibrium open system and an effective transmission factor is contained in the prefactor controlling the barrier crossing dynamics. As expected both are the functions of the external noise strength and the coupling of the noise to the bath modes. In the absence of external stochastic modulation, $\epsilon(t) = 0$, equation (3.18) reduces to standard Kramers' result [3], namely

$$K_{\text{kramers}} = \frac{\omega_0}{2\pi\omega_b}\left[\left\{\left(\frac{\beta}{2}\right)^2+\omega_b^2\right\}^{1/2} - \frac{\beta}{2}\right]\exp\left(\frac{-E_b}{k_BT}\right)$$

which can be verified with the explicit forms of the parameters D_0, B_0, D_b, B_b and Λ with $J_e = 0$, $I_e = \beta k_B T$, $g(x) = x$ and D or κ_0 equals to zero.

As the understanding of the theoretical aspects of nonequilibrium statistical mechanics became better a vast body of literature have emerged modifying Kramers' original approach which are well documented in the review by Hänggi, Talkner and Borkovec [4]. In the following we briefly discuss one of the issues from the post

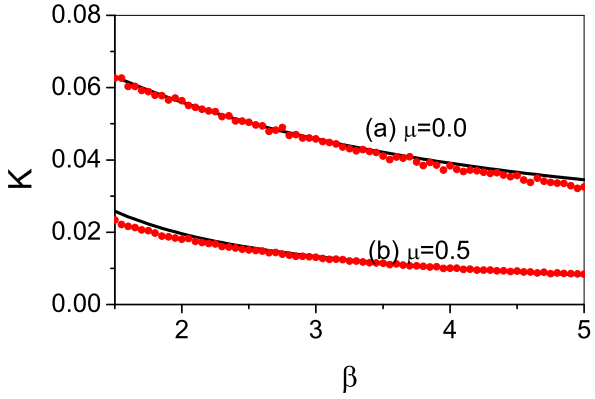

FIG. 1: (color online) Variation of escape rate K with dissipation constant β with (a) $\mu = 0$ and (b) $\mu = 0.5$ using the parameter set $b_1 = 1.0$, $E_b = 5.0$, $k_BT = 0.1$, $\kappa_0^2 = 5.0$, $D_e = 0.1$ and $\tau_e = 0.01$, where solid line and circle represents analytical and numerical rates, respectively.

Kramers development of the reaction rate theory which is closely connected to our work. Though Kramers' original approach was restricted by the Markovian assumption, however, in certain situations the memory effect becomes important and the generalized Langevin equation with a memory kernel must be accounted for. To the best of our knowledge following the seminal work in this direction by Grote and Hynes [30], Hänggi and Mojtabai [31] and Carmeli and Nitzan [32] have extended Kramers' work for an arbitrary memory friction and have found that the rate can often be larger than one would obtain from Kramers' original approach. The analysis of Hänggi and Mojtabai [31] was based on the non-Markovian Fokker-Planck equation of Adelman [34] for a parabolic poten-

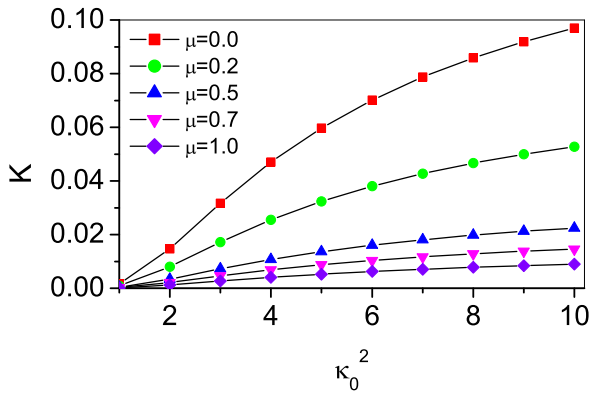

FIG. 2: (color online) Variation of analytical rate constant K with κ_0^2 for different values of the strength of nonlinearity μ with $b_1 = 1.0$, $E_b = 5.0$, $k_BT = 0.1$, $\beta = 1.0$, $D_e = 1.0$ and $\tau_e = 0.01$.

tial and they have used essentially the same approach of Kramers. The generalized Fokker-Planck approach have also been adopted by Carmeli and Nitzan [32] to derive the expression for the steady state escape rate in the non-Markovian regime. All the above mentioned theoretical approaches have been first supplemented with a full stochastic simulation by Straub,Borkovec and Berne [33] where the authors explicitly studied the dynamics within the framework of non-Markovian generalized Langevin equation. Incorporation of memory effect in the above mentioned work gets reflected in the rate expression

$$K_{\rm memory} = \frac{\omega_0}{2\pi\omega_b}\left[\left\{\left(\frac{\overline{\gamma}}{2}\right)^2 + \overline{\omega}_b^2\right\}^{1/2} - \frac{\overline{\gamma}}{2}\right]\exp\left(\frac{-E_b}{k_BT}\right)$$

where, $\overline{\gamma}$ and $\overline{\omega}_b$ are long time limit of the memory kernel $\gamma(t)$ and the renormalized frequency at the top of the potential $\omega_b(t)$, respectively. In this paper we have extended the above mentioned approaches [31, 32, 34] for state dependent diffusion in the non-Markovian regime to obtain a generalized steady state escape rate when the bath is modulated by an external stochastic force. Though our treatment is valid for small correlation time, it incorporates most of the characteristics of the non-Markovian state dependent diffusion process.

IV. SPECIFIC EXAMPLE: HEAT BATH DRIVEN BY EXTERNAL COLORED NOISE

As a specific example we consider that the heat bath is modulated externally by a colored noise $\epsilon(t)$ with noise correlation

$$\langle\epsilon(t)\epsilon(t')\rangle_e = \frac{D_e}{\tau_e}\exp\left(-\frac{|t-t'|}{\tau_e}\right) \qquad (4.1)$$

where D_e and τ_e are the strength and correlation time of the external noise respectively. In addition to that we consider the internal noise $f(t)$ to be white (*i.e.* $\tau_c \to 0$). The effective Gaussian-Ornstein-Uhlenbeck noise $\xi(t) = f(t) + \pi(t)$ will have an intensity D_R and a correlation time τ_R given by [16]

$$\begin{aligned} D_R &= \int_0^\infty \langle\xi(t)\xi(0)\rangle dt, \\ \tau_R &= \frac{1}{D_R}\int_0^\infty t\langle\xi(t)\xi(0)\rangle dt. \end{aligned}$$

Following the above definitions and using equation (2.9) we have (see equation (2.17)),

$$\begin{aligned} D_R &= c_0^2(k_BT + D_e\kappa_0^2) = \beta(k_BT + D_e\kappa_0^2), \quad (4.2a) \\ \tau_R &= \frac{D_e c_0^2\kappa_0^2}{D_R}\tau_e = \frac{\beta D_e\kappa_0^2}{D_R}\tau_e. \quad (4.2b) \end{aligned}$$

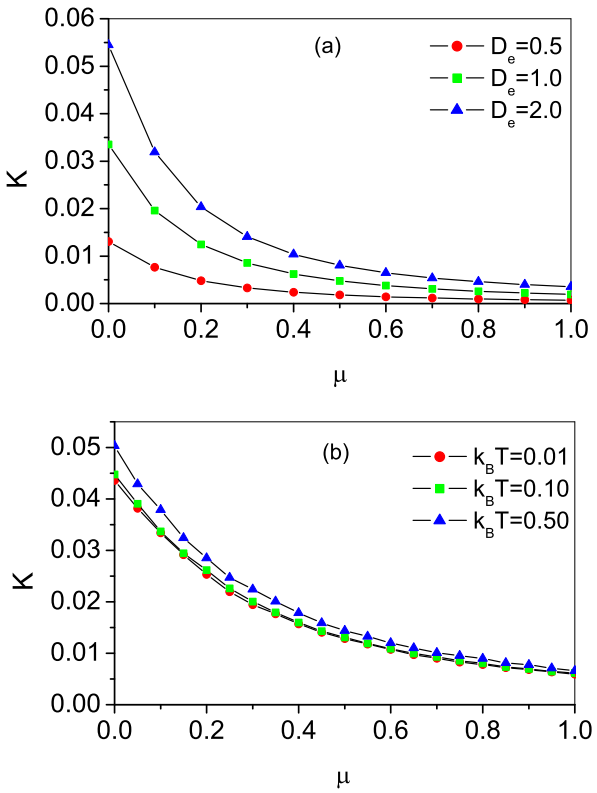

FIG. 3: (color online) Variation of rate constant K with strength of nonlinearity μ (a) for different values of external noise strength D_e (calculated analytically using $k_BT = 0.1$) and (b) for different values of temperature k_BT (calculated numerically using $D_e = 1.0$). The values of the other parameters used are $b_1 = 1.0$, $E_b = 5.0$, $\beta = 3.0$, $\kappa_0^2 = 5.0$ and $\tau_e = 0.01$.

Consequently the functions $A(x)$ and $B(x)$ in equation (3.4) becomes

$$\begin{aligned} A(x) &= (g'(x))^2 D_R - \Gamma(x)(g'(x))^2\tau_R D_R, \quad (4.3a) \\ B(x) &= (g'(x))^2\tau_R D_R. \quad (4.3b) \end{aligned}$$

From these equations we may evaluate the various parameters to obtain the generalized escape rate from equation (3.18).

V. RESULTS AND DISCUSSIONS

To study the dynamics we consider a model cubic potential of the form $V(x) = b_1x^2 - b_2x^3$ where b_1 and b_2 are the two constant parameters with $b_1, b_2 > 0$, so that the activation energy becomes $E_b = 4b_1^3/27b_2^2$. The nonlinear coupling function is taken to be $g(x) = x + (1/2)\mu x^2$, μ being a constant implying the strength of nonlinearity of the coupling function. We then numerically solve

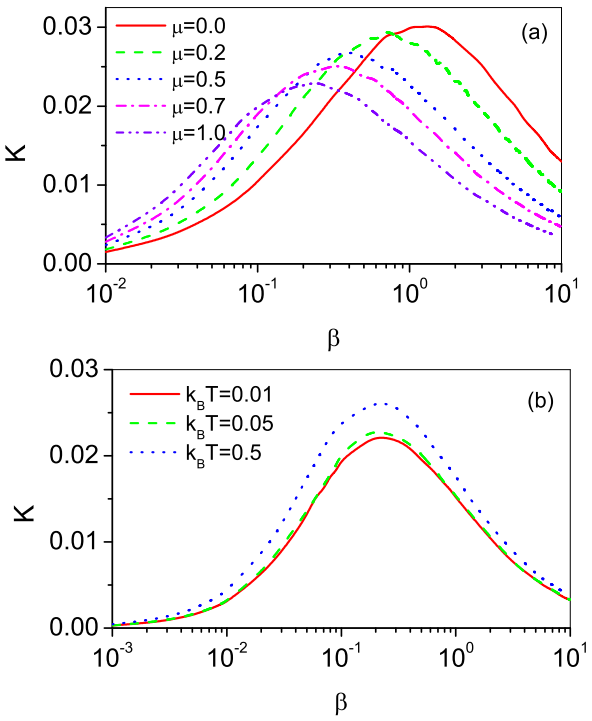

FIG. 4: (color online) Turnover of rate constant K (numerical) with dissipation constant β for (a) different values of μ (for $k_BT = 0.1$) and (b)different values of k_BT (for $\mu = 0.5$). The values of other parameters used are $b_1 = 1.0$, $E_b = 5.0$, $D_e = 1.0$, $\kappa_0^2 = 5.0$, $\tau_e = 1.0$.

the Langevin equation (3.2) using second order stochastic Heun's algorithm. To ensure the stability of our simulation we have used a small time step $\Delta t = 0.001$, with $\Delta t/\tau_R \ll 1$. The numerical rate has been defined as the inverse of the mean first passage time [35]. The mean first passage time has been calculated by averaging over $10,000$ trajectories. The value of other parameters used are given in the respective figure.

One of the result of Kramers' theory is that K varies inversely in the intermediate to strong damping regime. In Fig.1 we have plotted the rate constant K against the damping constant $\beta = c_0^2$ in the moderate to large damping region where our theory is valid and we compare the theoretical result (3.18) with the numerical simulation data for two different values of the nonlinear coupling parameter μ. It is observed that the agreement between the theoretical prediction and numerical simulation is quite satisfactory. In Fig.2 we plot the variation of rate constant K, obtained from theoretical result (3.18), with external coupling constant κ_0^2 for various nonlinearity parameters. We observe that for a given μ, the rate increases nearly linearly and for a particular value of κ_0, increase in μ causes decrease in rate, which is also observed from the Fig.3(a) (where we have plotted K vs. μ for different values of D_e from analytical result) and Fig.3(b) (where the same has been observed numerically for different temperatures).

In his dynamical theory of chemical reaction, Kramers identified two distinct regimes of stationary nonequilibrium states in terms of dissipation constant β. The essential result of Kramers' theory is that the rate varies linearly in the weak damping regime and inversely in the intermediate to strong damping regime. In between the two regimes the rate constant as a function of damping constant exhibits a bell-shaped curve known as Kramers' turnover [4]. In the traditional system reservoir model the dissipation and fluctuations are connected through the fluctuation-dissipation relation. A typical signature of this relation can been seen through this turnover phenomenon. Whereas for a thermodynamically open system where the heat bath is modulated by external noise, both the dissipation β and response function φ depend on the properties of the reservoir. Due to this connection between the dissipation and external noise source equation (2.9), plays the typical role of thermodynamic consistency relation, an analog of the fluctuation-dissipation relation for a thermodynamically closed system, for which one can expect turnover feature for this open system. In Fig.4(a) and Fig.4(b) we have plotted the rate constant K obtained from Langevin simulation for a wide range of damping constant for different values of nonlinearity parameter μ and temperature, respectively. The figures apart form demonstrating the turnover of the rate constant with the variation of the damping constant, shows a shifting of maxima towards left *i.e.*, weak damping regime with the increase of μ and also is consistent with Fig.3.

While observing the variation of the rate constant K as a function of correlation time (τ_e) of the external noise for different D_e (Fig.5(a)), different temperature (Fig.5(b)) and for different nonlinearity parameter μ (Fig.5(c)) we find an interesting result. In all cases K passes through a maxima, then decreases and ultimately becomes independent of τ_e for large values of τ_e. In short the rate of barrier crossing exhibits a resonance behavior with the correlation time of external noise which is responsible for fluctuation of barrier height. The above resonance phenomenon is known as resonant activation (RA) [36]. So far RA has been observed due to the barrier fluctuation as a result of direct driving of correlated noise to the system. In this model the barrier fluctuation occurs (3.2) due to the driving of nonlinearly coupled heat bath with the system by correlated noise. So the RA occurs as a result of correlated noise driven bath, which is interesting and new feature, instead of direct system driving. The immediate experimentally observable situation could be if we consider a simple unimolecular isomerization reaction in a photochemically active solvent under the influence of fluctuating light intensity (see section I), the reaction rate can be enhanced by tuning the correlation time of the fluctuating light field. It is also interesting to note that our nonlinear coupling model which yields a state dependent diffusion may have an important conse-

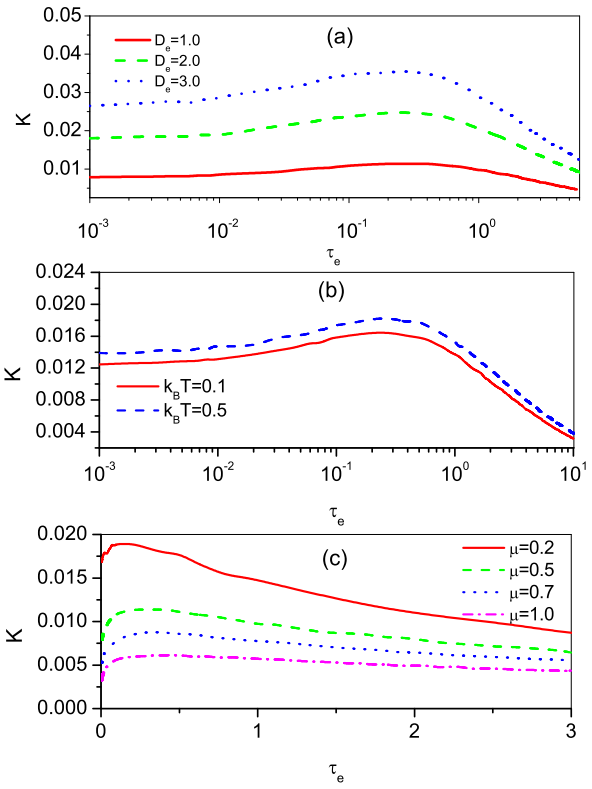

FIG. 5: (color online) Numerical variation of rate constant K with correlation of external noise τ_e for (a) different values of strength of external noise D_e (for $k_BT = 0.1$ and $\mu = 0.5$), (b) different values of k_BT (for $D_e = 1.0$ and $\mu = 0.5$) and (c) different values of μ (for $k_BT = 1.0$ and $D_e = 1.0$). The values of other parameters used are $b_1 = 1.0$, $E_b = 5.0$, $\beta = 5.0$ (for (b) $\beta = 3.0$) and $\kappa_0^2 = 5.0$. Note the logarithmic abscissa in (a) and (b).

quence in the generation of current for a Brownian particle moving in a periodic potential without any external bias. Because of its extraordinary success in explaining experimental observations on biomolecular motors active in muscle contractions, the state dependent diffusion has attracted wide attentions in recent years [37].

VI. CONCLUSION

Based on a system reservoir microscopic model where the system is nonlinearly coupled to a heat bath which is modulated by an external, stationary Gaussian noise with an arbitrary decaying correlation function, we have generalized the Kramers' theory and have numerically analyzed the model to calculate the steady state rate of escape from a metastable well. We have shown that since the reservoir is driven by the external noise and the dissipative properties of the system depend on the reservoir, a simple connection between the dissipation and the response function of the medium due to external noise can be established. We then followed the dynamics of the open system in the spatially-diffusion limited regime and derived the Fokker-Planck equation (corresponding to the Langevin equation with space dependent dissipation and multiplicative noise) with space dependent diffusion coefficient containing the effective temperature like quantity. We then derived the generalized Kramers' escape rate for moderate to strong damping regime. From the point of view of the realistic situation we considered the special case where the internal noise is white and the external noise is colored and have calculated the escape rate for a model cubic potential. We also numerically simulate the Langevin equation and observe that the theoretical prediction agrees reasonably well with the numerical result. Dependence of the rate K on various other parameters have been studied and it is observed that the enhancement of rate is possible by tuning the correlation time of external noise. The creation of a typical nonequilibrium open situation by modulating a bath with the help of an external noise is not an uncommon phenomena in applications and industrial processing. The external agency generating noise does work on the bath by stirring, pumping, agitating etc to which the system dissipates internally. In the present treatment we are concerned with a nonequilibrium steady state that signifies a constant throughput of energy. We believe that these considerations are likely to be important in other related issues in nonequilibrium open systems such as thermal ratchet and related problems.

So far, in this paper, we have considered a linear coupling in the interaction between the heat bath and the external driving noise, H_{int}. It will be interesting to see how the dynamics changes when H_{int} is nonlinear. Studies have been made recently on anomalous diffusion in presence of correlated external noise [38]. Our present methodology may be extended to investigate the transport process when the bath is modulated by two correlated external noise. In our future communication we would like to address such issues.

Acknowledgments

We are thankful to Professor D.S. Ray for stimulating discussions and critical comments. JRC is also thankful to Dr. B. Deb for constant encouragement and to Professor J.K. Bhattacharjee for useful discussions. JRC is indebted to Indian Academy of Science for providing a summer fellowship. SKB acknowledges support from Virginia Tech through ASPIRES award program.

APPENDIX A: DERIVATION OF FOKKER-PLANCK EQUATION

Equation (3.2) can be written in the following form

$$\dot{u_1} = G_1(u_1, u_2, t; f(t), \pi(t)) \quad \text{(A1a)}$$
$$\dot{u_2} = G_2(u_1, u_2, t; f(t), \pi(t)) \quad \text{(A1b)}$$

where we have used the following abbreviations

$$u_1 = x \quad \text{(A2a)}$$
$$u_2 = v \quad \text{(A2b)}$$

and

$$G_1 = v \quad \text{(A3a)}$$
$$G_2 = -\frac{dV(x)}{dx} - \Gamma(x)v + g'(x)\{f(t) + \pi(t)\} \quad \text{(A3b)}$$

The vector u with components u_1 and u_2 thus represents a point in a two-dimensional "phase space" and the equation (A1a-A1b) determines the velocity at each point in this phase space. The conservation of the points now asserts the following linear equation of motion for density $\rho(u,t)$ in phase space [29]

$$\frac{\partial}{\partial t}\rho(u,t) = -\sum_{n=1}^{2}\frac{\partial}{\partial u_n}G_n(u,t,f(t),\pi(t))\rho(u,t)$$

or more compactly

$$\frac{\partial \rho}{\partial t} = -\nabla \cdot G\rho. \quad \text{(A4)}$$

Our next task is to find out a differential equation whose average solution is given by $\langle\rho\rangle$ [29] where the stochastic averages has to be performed over two noise processes $f(t)$ and $\epsilon(t)$. $\nabla \cdot G$ can be partitioned into two parts: a constant part $\nabla \cdot G_0$ and a fluctuating part $\nabla \cdot G_1(t)$, containing these noises. Thus we write

$$\nabla \cdot G(u,t,f(t),\pi(t)) = \nabla \cdot G_0(u) + \alpha \nabla \cdot G_1(u,t,f(t),\pi(t)) \quad \text{(A5)}$$

where α is a parameter (we put it as an external parameter to keep track of the order of the perturbation expansion in $\alpha\tau_e$, τ_e is the correlation time of the $\epsilon(t)$, we put $\alpha = 1$ at the end of the calculation) and also note that $\langle\langle G_1(t)\rangle\rangle = 0$. Equation (A4) therefore takes the following form

$$\dot{\rho}(u,t) = (A_0 + \alpha A_1)\rho(u,t) \quad \text{(A6)}$$

where $A_0 = -\nabla \cdot G_0$ and $A_1 = -\nabla \cdot G_1$. The symbol ∇ is used for the operator that differentiate everything that comes after it with respect to u. Making use of one of the main results for the theory of linear equation of the form (A6) with multiplicative noise [29], we derive an average equation for ρ [$\langle\rho\rangle = P(u,t)$, the probability density of $u(t)$]

$$\frac{\partial P}{\partial t} = \left\{A_0 + \alpha^2 \int_0^\infty d\tau \langle A_1(t)\exp(\tau A_0)A_1(t-\tau)\rangle \times \exp(-\tau A_0)\right\} P. \quad \text{(A7)}$$

The above result is based on second order cumulant expansion and is valid for the rapid fluctuations with small strength where the correlation time τ_e is short but finite [29], *i.e.*,

$$\langle A_1(t)A_1(t')\rangle = 0 \text{ for } |t-t'| > \tau_e.$$

Equation (A7) is exact in the limit τ_e trends to zero. Using the expansions for A_0 and A_1 we obtain

$$\frac{\partial P}{\partial t} = \left\{-\nabla \cdot G_0 + \alpha^2 \int_0^\infty d\tau \langle \nabla \cdot G_1(t)\exp(-\tau \nabla \cdot G_0) \times \nabla \cdot G_1(t-\tau)\rangle \exp(\tau \nabla \cdot G_0)\right\} P. \quad \text{(A8)}$$

The operator $\exp(-\tau \nabla \cdot G_0)$ in the above equation provides the solution of the equation

$$\frac{\partial \mathscr{G}(u,t)}{\partial t} = -\nabla \cdot G_0 \mathscr{G}(u,t) \quad \text{(A9)}$$

($\mathscr{G}$ signifies the unperturbed part of ρ) which can be found explicitly in terms of characteristic curves. The equation

$$\dot{u} = G_0(u) \quad \text{(A10)}$$

for fixed t determines a mapping from $u(\tau = 0)$ to $u(\tau)$, *i.e.*, $u \to u^\tau$ with the inverse $(u^\tau)^{-\tau} = u$. The solution of equation (A9) is given by

$$\mathscr{G}(u,t) = \mathscr{G}(u^{-t},0)\left|\frac{d(u^{-t})}{d(u)}\right| = \exp(-t\nabla \cdot G_0)\mathscr{G}(u,0), \quad \text{(A11)}$$

$|d(u^{-t})/d(u)|$ being a Jacobian determinant. The effect of $\exp(-t\nabla \cdot G_0)$ on $\mathscr{G}(u)$ is given by

$$\exp(-t\nabla \cdot G_0)\mathscr{G}(u,0) = \mathscr{G}(u^{-t},0)\left|\frac{d(u^{-t})}{d(u)}\right|. \quad \text{(A12)}$$

The above simplification when we put in equation (A8) yields

$$\frac{\partial P}{\partial t} = \nabla\cdot\left\{-G_0+\alpha^2\int_0^\infty d\tau\left|\frac{d(u^{-\tau})}{d(u)}\right| \times\langle G_1(u,t)\nabla_{-\tau}\cdot G_1(u^{-\tau},t-\tau)\rangle \times\left|\frac{d(u)}{d(u^{-\tau})}\right|\right\}P. \quad \text{(A13)}$$

where $\nabla_{-\tau}$ denotes differentiation with respect to $u_{-\tau}$. We put $\alpha = 1$ for the rest of the treatment. We now identify

$$\begin{aligned} u_1 &= x \\ u_2 &= v \\ G_{01} &= v,\ G_{11}=0 \\ G_{02} &= -\Gamma(x)v - V'(x) \\ G_{12} &= g'(x)\{f(t)+\pi(t)\}. \end{aligned} \quad \text{(A14)}$$

In this notation equation (A13) now reduces to

$$\begin{aligned} \frac{\partial P}{\partial t} &= -\frac{\partial}{\partial x}(vP)+\frac{\partial}{\partial v}\{\Gamma(x)v+V'(x)\}P \\ &+\frac{\partial}{\partial v}\int_0^\infty d\tau\langle[g'(x)\{f(t)+\pi(t)\}][\frac{\partial}{\partial v^{-\tau}} \\ &\times\{g'(x^{-\tau})(f(t-\tau)+\pi(t-\tau))\}]\rangle P \end{aligned} \quad \text{(A15)}$$

where we have used the fact that the Jacobian obeys the equation [29]

$$\frac{d}{dt}\log\left|\frac{d(x^t,v^t)}{d(x,v)}\right| = \frac{\partial v}{\partial x}+\frac{\partial}{\partial v}\{-\Gamma v+V'(x)\} = -\Gamma,$$

so that the Jacobian equals to $e^{-\Gamma t}$.

As a next approximation we consider the"unperturbed" part of equation (A1a-A1b) and take the variation of v during τ_e into account to first order in τ_e. Thus we have

$$x^{-\tau} = x-\tau v,\ v^{-\tau} = v+\Gamma\tau v+\tau V'(x). \quad \text{(A16)}$$

Neglecting terms $\mathscr{O}(\tau^2)$, equation (A16) yields

$$\frac{\partial}{\partial v^{-\tau}} = (1-\Gamma\tau)\frac{\partial}{\partial v}+\tau\frac{\partial}{\partial x}.$$

Taking this into consideration equation (A15) can be simplified after some algebra to the following form

$$\begin{aligned} \frac{\partial P(x,v,t)}{\partial t} &= -v\frac{\partial P}{\partial x} \\ &+[\Gamma(x)v+V'(x)-2g'(x)g''(x)J_e]\frac{\partial P}{\partial v} \\ &+A\frac{\partial^2 P}{\partial v^2}+B\frac{\partial^2 P}{\partial v\partial x}+\Gamma(x)P \end{aligned} \quad \text{(A17)}$$

where

$$A = (g'(x))^2 I_e - \Gamma(x)(g'(x))^2 J_e \ \text{ and } B = (g'(x))^2 J_e, \quad \text{(A18)}$$

and I_e and J_e are defined as

$$I_e = \int_0^\infty \langle\xi(t)\xi(t-\tau)\rangle d\tau, \quad \text{(A19a)}$$

$$J_e = \int_0^\infty \tau\langle\xi(t)\xi(t-\tau)\rangle d\tau. \quad \text{(A19b)}$$

APPENDIX B: DERIVATION OF ESCAPE RATE

Inserting (3.16) in (3.14), we obtain the equation for $F(x,v)$ in the steady state in the neighborhood of x_b

$$\begin{aligned} &-\ (1+B_b/D_b)v\frac{\partial F}{\partial x}-\left[\frac{D_b}{D_b+B_b}\omega_b^2(x-x_b)\right. \\ &+\ \left.\Gamma(x_b)v\right]\frac{\partial F}{\partial v}+A_b\frac{\partial^2 F}{\partial v^2}+B_b\frac{\partial^2 F}{\partial v\partial x}=0. \end{aligned} \quad \text{(B1)}$$

At this point we set

$$u = v+a(x-x_b), \quad \text{(B2)}$$

where a is a constant to be determined. With the help of transformation (B2), equation (B1) reduces to the following form

$$\begin{aligned} &\{A_b+aB_b\}\frac{d^2F}{du^2}-\left[\frac{D_b}{D_b+B_b}\omega_b^2(x-x_b)\right. \\ &+\left.\left\{\Gamma(x_b)+a\left(1+\frac{B_b}{D_b}\right)\right\}v\right]\frac{dF}{du}=0. \end{aligned} \quad \text{(B3)}$$

Now let

$$\frac{D_b}{D_b+B_b}\omega_b^2(x-x_b)+\left\{\Gamma(x_b)+a\left(1+\frac{B_b}{D_b}\right)\right\}v = -\lambda u \quad \text{(B4)}$$

where λ is another constant to be determined later. By virtue of the relation (B4), equation (B3) becomes

$$\frac{d^2F}{du^2}+\Lambda u\frac{dF}{du}=0 \quad \text{(B5)}$$

where

$$\Lambda = \frac{\lambda}{A_b+aB_b}. \quad \text{(B6)}$$

The two constants λ and a must satisfy the simultaneous relations

$$-\lambda a = \frac{D_b}{D_b+B_b}\omega_b^2,$$
$$-\lambda = \Gamma(x_b) + a\left(1+\frac{B_b}{D_b}\right).$$

This implies that the constant a must satisfy the quadratic equation

$$\frac{D_b+B_b}{D_b}a^2 + \Gamma(x_b)a - \frac{D_b}{D_b+B_b}\omega_b^2 = 0$$

which allows the solutions for a as

$$a_\pm = \frac{D_b}{2(D_b+B_b)}\left\{-\Gamma(x_b) \pm \sqrt{\Gamma^2(x_b)+4\omega_b^2}\right\}. \quad \text{(B7)}$$

The general solution of equation (B5) is

$$F(u) = F_2\int_0^u \exp\left(-\frac{\Lambda z^2}{2}\right)dz + F_1, \quad \text{(B8)}$$

where F_1 and F_2 are constant of integration. We look for a solution which vanishes for large x. This condition is satisfied if the integration in (B8) remain finite for $|u| \to +\infty$. This implies that $\Lambda > 0$ so that only a_- becomes relevant. Then the requirement $P_b(x,v) \to 0$ for $x \to +\infty$ yields

$$F_1 = F_2\sqrt{\pi/2\Lambda}. \quad \text{(B9)}$$

Thus we have

$$F(u) = F_2\left[\sqrt{\frac{\pi}{2\Lambda}} + \int_0^u \exp\left(-\frac{\Lambda z^2}{2}\right)dz\right]$$

and correspondingly

$$P_b(x,v) = F_2\left[\sqrt{\frac{\pi}{2\Lambda}} + \int_0^u \exp\left(-\frac{\Lambda z^2}{2}\right)dz\right] \times \exp\left[-\left\{\frac{v^2}{2D_b} + \frac{V(x)}{D_b+B_b}\right\}\right]. \quad \text{(B10)}$$

The current across the barrier associated with the steady state distribution is given by

$$j = \int_{-\infty}^{+\infty} vP_b(x=x_b,v)dv$$

which may be evaluated using (B10) and the linearized version of $V(x)$, namely $V(x) = E_b - (1/2)\omega_b^2(x-x_b)^2$ as

$$j = F_2\left(\frac{2\pi}{\Lambda + D_b^{-1}}\right)^{1/2} D_b \exp\left(\frac{-E_b}{D_b+B_b}\right). \quad \text{(B11)}$$

To determine the remaining constant F_2 we proceed as follows. We first note that as $x \to -\infty$ the pre-exponential factor in equation (B10) reduces to the following form

$$F_2[...] = F_2\left(\frac{2\pi}{\Lambda}\right)^{1/2}. \quad \text{(B12)}$$

We then obtain the reduced distribution function in x as

$$\widetilde{P}_b(x\to-\infty) = 2\pi F_2\left(\frac{D_b}{\Lambda}\right)^{1/2}\exp\left(\frac{-V(x)}{D_b+B_b}\right), \quad \text{(B13)}$$

where we have used the definition for the reduced distribution as

$$\widetilde{P}(x) = \int_{-\infty}^{+\infty} P(x,v)dv.$$

Similarly we derive the reduced distribution in the left well around $x \approx 0$ using equation (3.11) where the linearized potential is $V(x) = \omega_0^2x^2/2$,

$$\widetilde{P}_{st}(x) = \frac{1}{Z}\sqrt{2\pi D_0}\exp\left(\frac{-\omega_0^2x^2}{2(D_0+B_0)}\right) \quad \text{(B14)}$$

with the normalization constant $1/Z$ given by

$$\frac{1}{Z} = \frac{\omega_0}{2\pi\sqrt{D_0(D_0+B_0)}}.$$

The comparison of the distribution (B13) and (B14) near $x \approx 0$, *i.e.*,

$$\widetilde{P}_{st}(x_0) = \widetilde{P}_b(x_0)$$

gives

$$F_2 = \left(\frac{\Lambda}{D_b}\right)^{1/2}\frac{\omega_0}{2\pi\sqrt{2\pi(D_0+B_0)}}. \quad \text{(B15)}$$

Hence from equation (B11), the normalized current or the barrier crossing rate K, for moderate to large friction, is given by

$$K = \frac{\omega_0}{2\pi}\frac{D_b}{(D_0+B_0)^{1/2}}\left(\frac{\Lambda}{1+\Lambda D_b}\right)^{1/2}\exp\left(\frac{-E_b}{D_b+B_b}\right) \quad \text{(B16)}$$

where E_b is the potential barrier height.

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